Ultrafast Carrier Dynamics and Third Generation Photovoltaics

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A critical aspect in reducing the module cost for photovoltaic systems is improvement of the solar cell efficiencies beyond the single bandgap limit of approximately 32%. This limit is a consequence of the fact that each absorbed photon contributes only the energy of the electron-hole pair it generates, independent of the photon energy (the excess energy is eventually lost to phonons). So called third generation concepts circumvent this limit for example generation of multiple electron-hole pairs from a single photon through multiexciton generation.[1] Another approach is that of hot carrier solar cells,[2] in which carriers are extracted from energy selective contacts before losing their excess kinetic energy to the lattice. In both cases, the dynamics of carrier relaxation in terms of electron-phonon interaction are of crucial importance in realizing quantum efficiencies much greater than unity. Particularly promising are the nanocrystalline materials, where the reduced dimensionality of the system suppresses the dominant optical phonon relaxation mechanisms.

Here we investigate the short time carrier relaxation in multiexciton and hot electron solar cells under varying photoexcitation conditions using ensemble Monte Carlo (EMC) simulation coupled with rate equation and thermodynanmic models, to understand the limiting factors affecting 3G cell performance. The EMC code simulates the dynamics of photoexcited electrons and holes in the quantum confined states in III-V nanostructure systems. The initial and final state transition rates are calculated between the quantized states of the system, giving rise to both intrasubband and intersubband processes in terms of carrier relaxation. Scattering processes due to optical phonons (polar and nonpolar), dissipative acoustic phonons, and ionized impurities are included. Electron-electron, electron-hole, and hole-hole scattering between quantum confined states within a screened, multi-subband framework, described in detail elsewhere.[3,] Hot phonons may play a critical role in reducing carrier energy loss, and maintaining energy within the absorber system.[4] Here we explicitly include nonequilibrium optical phonons, in which the anharmonic decay of the optical phonon population to acoustic phonons is described using a phenomenological phonon lifetime. Here we find that the optical phonon lifetime plays a critical role in the performance of e.g. hot carrier solar cells, where decay times in excess of several 10s of picoseconds are required to realize high performance in such systems.

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3 S. M. Goodnick and P. Lugli, *Hot Carrier relaxation in quasi-2D systems*, in Hot Carriers in Semiconductor Microstructures: Physics and Applications, (J. Shah, Ed.), Academic Press Inc., pp. 191-234, 1992.

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